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Citation: AIP Conference Proceedings **1501**, 247 (2012); doi: 10.1063/1.4769515
View online: http://dx.doi.org/10.1063/1.4769515
View Table of Contents: http://scitation.aip.org/content/aip/proceeding/aipcp/1501?ver=pdfcov
Published by the AIP Publishing

Heat and Mass Transfer in Gases due to Pressure and Temperature Gradients in a Laser Radiation Field

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Abstract. Heat and mass transfer in a one-component gas through a capillary in the field of resonant laser radiation in the presence of pressure and temperature gradients are considered. On the basis of the Boltzmann type kinetic equations in the linear approximation the expression for entropy production is obtained. Kinetic coefficients satisfy the Onsager reciprocity relations at all Knudsen numbers and for any nature of the interaction of gas atoms with the surface of the capillary. The pressure and temperature gradients established in the insulated system in a laser field are defined in a nearly free molecular regime.

Keywords: one-component gas, optical radiation, kinetic equations, transport phenomena, entropy production, kinetic coefficients
PACS: 47.45.Ab, 51.10.+y

INTRODUCTION

The phenomena of light-induced drift (LID) and heat transfer (LIHT) occur in the gas consisting of atoms and molecules absorbing selectively the monochromatic light according to their velocities [1]. The feature of the laser gas-kinetics is that the gradients of the thermodynamic parameters are not specified, but they are result of light-induced transport phenomena. These gradients in further stimulate the normal heat and mass transfer in the gas.

The problem is that the LID and LIHT can not be measured directly. LID is investigated experimentally measuring the pressure gradient in a closed capillary [2]. Probably, the LIHT can be investigated experimentally by measurement of the temperature gradient, which is established in the insulated system. In this context, it seems interesting to construct a theory that establishes the relationship between the characteristics of resonance radiation on the one hand, and the gradients of temperature and pressure on the other.

In [3], transport phenomena in a mixture of optically active and buffer gases in a laser radiation field are considered. The gradients of pressure, temperature and concentration are added as a source of nonequilibrium. On the basis of the fundamental properties of kinetic equations and the gas-surface interactions Onsager reciprocity relations for kinetic coefficients are proved. However, the proof is based on the few reasonable assumptions. In particular, the possibility of radiative decay of the ground state of atoms is allowed. Assumptions relating to the relaxation of the population of ground and excited levels are the consequence of the independence of the kinetic coefficients on the frequency of the radiative decay of the excited level. This contradicts the result of [4], which is obtained by direct numerical solution of kinetic equations.

The purpose of this paper is to analyze the processes of heat and mass transfer in a gas through a capillary in the presence of pressure and temperature gradients under the laser field. The expressions for the entropy production and kinetic coefficients satisfying Onsager's reciprocity for all Knudsen numbers (Kn) and arbitrary nature of the gas-surface interactions are given in the report.

Estimates of pressure and temperature gradients, established in a closed insulated system under resonant laser radiation are presented for almost free-molecular regime.

STATEMENT OF THE PROBLEM

Consider a one-component gas confined in a circular-cylindrical tube of radius r_0. A traveling light wave is propagating along the axis z coinciding with the axis of the capillary. Pressure and temperature gradients are directed along the same axis (Fig.1). The gas particles are assumed to be in two quantum state-ground n and excited m. The radiation frequency ω is slightly detuned from the center of the absorption line at ω_{mn}, i.e. $\Omega = \omega - \omega_{mn}$ $\left(|\Omega| << \omega, \omega_{mn}\right)$. Atoms that have absorbed radiation change the kinetic cross section. Simultaneously with the stimulated transitions there is a competitive process-radiative decay of the excited level with frequency Γ_m. Thus, the gas phase can be interpreted as a binary mixture consisting of the

28th International Symposium on Rarefied Gas Dynamics 2012
AIP Conf. Proc. 1501, 247-253 (2012); doi: 10.1063/1.4769515

particles with equal masses m but different cross sections. Stimulated transitions and radioactive decay of the excited levels lead to the exchange of atoms between excited and unexcited components. It leads to the appearance of light-induced heat and mass flows of excited and unexcited particles in the channel and, under certain conditions, there are a drift of the gas as a whole (LID) and heat flux (LIHT) [5]. Light-induced currents are superimposed on the flows caused by pressure and temperature gradients that lead to the emergence of new cross-effects.

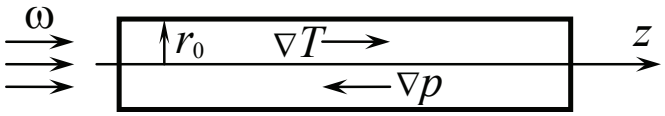

FIGURE 1. Geometry of the problem.

The state of the gas mixture can be described by the distribution functions for the excited f_m and unexcited f_n atoms, which satisfy the following Boltzmann kinetic equations [6]:

$$\frac{\partial f_n}{\partial t}+\mathbf{v}_\perp\frac{\partial f_n}{\partial \mathbf{r}}+v_z\frac{\partial f_n}{\partial z}=\frac{\Gamma_m\chi(\mathbf{v})}{2}\left(f_m-f_n\right)+\Gamma_m f_m+S_n\,, \tag{1}$$

$$\frac{\partial f_m}{\partial t}+\mathbf{v}_\perp\frac{\partial f_m}{\partial \mathbf{r}}+v_z\frac{\partial f_m}{\partial z}=\frac{\Gamma_m\chi(\mathbf{v})}{2}\left(f_n-f_m\right)-\Gamma_m f_m+S_m\,,$$

$$\chi(\mathbf{v})=\frac{4\left|G_{mn}\right|^2\Gamma}{\Gamma_m\left[\Gamma^2+\left(\Omega-\mathbf{kv}\right)^2\right]},\quad G_{mn}=\frac{Ed_{mn}}{2\hbar}.$$

Here Γ_m is the rate of radiative decay of the excited level, Γ is the homogeneous half-width of absorption line of molecular transition *m-n*, E is the amplitude of the electrical field in the light wave, d_{mn} is the dipole matrix element of m-n transition, $\mathbf{k}$ -wave vector, $\mathbf{v}_\perp,\mathbf{r}$ are two-dimensional vectors in the cross-section of the capillary, $\hbar$ is the Planck constant, S_n and S_m are Boltzmann collision integrals of atoms in a ground and excited state respectively, G_{mn} is the Rabi frequency; $\chi(\mathbf{v})$ is the probability of absorption per unit time for atoms with a given velocity $\mathbf{v}$.

The boundary conditions for the distribution functions f_i with arbitrary scattering kernel $R\left(\mathbf{v}'\to\mathbf{v}\right)$ have the form [7]:

$$\left|\mathbf{vn}\right|f_i^+(\mathbf{v})=\int\limits_{(\mathbf{v}'\mathbf{n})<0}R\left(\mathbf{v}'\to\mathbf{v}\right)f_i^-(\mathbf{v}')\left|\mathbf{v}'\mathbf{n}\right|d\mathbf{v}'\,;\quad i=m,n\,;\ (\mathbf{vn})>0\,, \tag{2}$$

Here f_i^+, f_i^- – the distribution function of reflected and incident on the surface atoms, respectively.

The distribution functions of excited and unexcited atoms in the weak-field approximation and small gradients of the thermodynamic parameters are slightly different from the equilibrium Maxwell-Boltzmann distributions:

$$f_i=f_{i0}\left[1+h_i(\mathbf{r},\mathbf{v})\right],\qquad \left\|h_i\right\|<<1 \tag{3}$$

$$f_{i0}=n_i(z)\left(\frac{m}{2\pi k_BT(z)}\right)^{3/2}\exp\left(-\frac{mv^2}{2k_BT(z)}\right),$$

$$n_i(z) = n(z)\frac{\exp(-E_i/k_BT(z))}{\sum_{i=n,m}\exp(-E_i/k_BT(z))}, \qquad i = n,m\,.$$

Here E_i is internal energy of the atom in *i*-th state, k_B is the Boltzmann constant, $T(z)$, $n(z)$ and $n_i(z)$ are the local values of the gas temperature, the total number density and the population of the i-th level.

Kinetic equations (1), linearized with respect to perturbation functions h_i, take the following form:

$$\frac{\partial h_i}{\partial t} + \boldsymbol{v}_\perp \frac{\partial h_i}{\partial \boldsymbol{r}} = \sum_{j=m,n} L_{ij}(h_i) + R_i h_m + \frac{\overline{v}}{r_0} g_i\,, \tag{4}$$

where

$$g_i = -c_z\nu - c_z\left(c^2 - \frac{5}{2} + \varepsilon_i - \overline{\varepsilon}\right)\tau + \frac{\Gamma_m \chi(v_z) r_0}{2\overline{v}}\left(\frac{n_j}{n_i} - 1\right), \qquad i,j = n,m\,;\ i \neq j$$

$$\nu = \frac{r_0}{p}\frac{\partial p}{\partial z}, \qquad \tau = \frac{r_0}{T}\frac{\partial T}{\partial z}, \qquad c_i = \frac{v_i}{\overline{v}}, \qquad c^2 = c_z^2 + c_\perp^2,$$

$$\overline{\varepsilon} = \frac{\sum_{i=n,m}\varepsilon_i \exp(-\varepsilon_i)}{\sum \exp(-\varepsilon_i)}, \qquad R_i = \begin{cases} \Gamma_m\left(\dfrac{n_m}{n_n}\right), i = n \\ -\Gamma_m, i = m \end{cases}, \qquad \varepsilon_i = \frac{E_i}{k_BT_0}, \qquad \overline{v} = \left(\frac{2k_BT_0}{m}\right)^{1/2};$$

$L_{ij}(h_i)$ – linearized collision integral.

THE ENTROPY PRODUCTION

The entropy of the gas mixture in a volume *V* per unit length of the capillary is defined as

$$S_e = -k_B \sum_{i=m,n}\int_V dV \int f_i \ln f_i d\mathbf{v}\,. \tag{5}$$

Using the linearization (3), we have

$$\frac{\partial S_e}{\partial t} = -k_B \sum_i \int_V dV \int f_{i0} h_i \frac{\partial h_i}{\partial t} d\boldsymbol{v} = \sigma - \frac{\overline{v}}{r_0} k_B \sum_{i=m,n}\int_V dV \int f_{i0} h_i g_i d\boldsymbol{v}\,, \tag{6}$$

where σ is the entropy production. The last equality in (6) is obtained by substituting the expression $\partial h_i / \partial t$ from Eq. (4). The entropy production is determined by three processes:

- due to intermolecular collisions $\sigma_{coll} \geq 0$ [8]

$$\sigma_{coll} = -k_B \sum_{(i,k)=n,m}\int_V dV \int f_{io} h_i L_{ik}(h_i) d\mathbf{v}\ ; \tag{7}$$

- due to the spontaneous decay of the excited level $\sigma_R \geq 0$

$$\sigma_R = -k_B \sum_i \int_V dV \int f_{i0} h_i h_m R_i d\boldsymbol{v} = k_B \Gamma_m \int_V dV \int f_{m0}\left(h_m^2 - h_n h_m\right) d\boldsymbol{v}\ ; \tag{8}$$

- due to the interaction of atoms with boundary surface $\sigma_\Sigma \geq 0$ [7]:

$$\sigma_\Sigma = k_B \int_V dV \sum_{i=n,m} \int f_{i0} h_i \mathbf{v}_\perp (\nabla h_i) d\mathbf{v} = k_B \sum_{i=n,m} \int_\Sigma d\Sigma \int (\mathbf{n}\mathbf{v}_\perp) f_{i0} \frac{h_i^2}{2} d\mathbf{v}, \tag{9}$$

where Σ is the surface area of the volume V. Consequently, the entropy production is $\sigma = \sigma_{coll} + \sigma_R + \sigma_\Sigma \geq 0$.

KINETIC COEFFICIENTS

Stationary and weakly nonequilibrium state of the gas caused by the small constant gradients $(\nu, \tau << 1)$ is considered. Based on the type of expression for the entropy production the generalized forces are chosen as follows:

$$X_p = -k_B \nu \ , \quad X_T = -\frac{\tau}{T_0}, \quad X_S = \chi_0 = \int_{-\infty}^{+\infty} \chi(c_z) e^{-c_z^2} dc_z \ . \tag{10}$$

The first two forces for non-isothermal gas flow in the channel were used in [9], and the force X_S is associated with a resonant optical radiation. Choice force X_S in the form (10) is useful, since χ_0 determines the probability to absorb the radiation per unit time by particles [10]. The quantity of X_S is measured in the optical spectral experiments; it is proportional to the intensity of radiation I. Force X_S is independent of the sign of the detuning Ω and it reaches maximum value at the exact resonance $\Omega = 0$.

In the steady state we have $\partial S_e / \partial t = 0$. Then the entropy production (6) is expressed as

$$\sigma = \frac{\overline{v}}{r_0} k_B \sum_i \int_S dS \int f_{i0} h_i g_i d\boldsymbol{\nu} = \sum_k J_k X_k \ , \quad k = p, T, S \ . \tag{11}$$

Here

$$J_p = (\varphi_p, h_i), \quad J_T = k_B T_0 (\varphi_T, h_i), \quad J_S = k_B (\varphi_S, h_i),$$

$$(\varphi_l, h_i) = \sum_i \int_S dS \int \varphi_i f_{i0} h_i d\mathbf{v}, \quad l = p, T, S, \tag{12}$$

$$\varphi_p = \frac{c_z \overline{v}}{r_0}, \quad \varphi_T = \frac{c_z \overline{v}}{r_0}\left(c^2 - \frac{5}{2}\right), \quad \varphi_S = \Gamma_m \frac{\chi(c_z)}{2\chi_0}\left(\frac{n_j}{n_i} - 1\right), \quad i, j = n, m \, ; \, i \neq j,$$

S is the cross-sectional area of the capillary; J_p, J_T are the densities of the gas flow and heat flux respectively, J_S is the density of entropy flux.

The perturbation functions in the linear approximation can be written as

$$h_i = h_i^p \nu + h_i^T \tau + h_i^S \chi_0 \ . \tag{13}$$

Substituting (13) into (12), we obtain the following expressions for the fluxes

$$J_l = \sum_k \Lambda_{lk} X_k \ , \quad k, l = p, T, S. \tag{14}$$

The kinetic coefficients have the form

$$\Lambda_{pp} = -\frac{1}{k_B}(\varphi_p, h_i^p), \quad \Lambda_{pT} = -T_0 (\varphi_p, h_i^T), \quad \Lambda_{pS} = (\varphi_p, h_i^S),$$

$$\Lambda_{Tp} = -T_0\left(\varphi_T, h_i^p\right), \qquad \Lambda_{TT} = -k_B T_0^2\left(\varphi_T, h_i^T\right), \qquad \Lambda_{TS} = k_B T_0\left(\varphi_T, h_i^S\right), \tag{15}$$

$$\Lambda_{Sp} = -\left(\varphi_S, h_i^p\right), \qquad \Lambda_{ST} = -k_B T_0\left(\varphi_S, h_i^T\right), \qquad \Lambda_{SS} = k_B\left(\varphi_S, h_i^S\right).$$

Here Λ_{lk} - Onsager kinetic coefficients, which determine the contribution of the various thermodynamic forces in streams. According to (14), the direct and cross-currents processes take place into the system. Thus, the pressure gradient causes Poiseuille flow (Λ_{pp}), the temperature gradient leads to a heat flux (Λ_{TT}), and the force X_S causes the flow of entropy along the capillary due to energy transfer radiation (Λ_{SS}). The forces X_k can cause cross-currents J_i ($i \neq k$), which are characterized by the kinetic coefficients Λ_{lk} . The temperature gradient can stimulate the flow of gas (thermal creep Λ_{pT}) and flow of entropy (thermalentropic effect Λ_{ST}), and the pressure gradient causes the flow of heat (mechanocaloric effect Λ_{Tp}) and the entropy flux (baroentropic effect Λ_{Sp}). At the same time, the impact of laser radiation on the atoms leads to a gas drift (Λ_{pS}) and heat transfer (Λ_{TS}), which are cross-phenomena in relation to the baroentropic and thermalentropic effects, respectively.

In discontinuous systems for cross-coefficient according to the thermodynamics of nonequilibrium processes are postulated Onsager reciprocity relations:

$$\Lambda_{lk} = \Lambda_{kl} \ . \tag{16}$$

The symmetry of the Onsager matrix (16) is proved in an obvious way. Substituting (13) in Eq. (4) and choosing terms with the same generalized forces, we obtain a system of kinetic equations which implies the following relations:

$$h_i^p\left(c^2 - \frac{5}{2}\right) = h_i^T, \quad h_i^p \varphi_S = -\varphi_p h_i^S, \quad h_i^T \varphi_S = -\varphi_T h_i^S. \tag{17}$$

In view of (17) expression (15) for the cross kinetic coefficients satisfy the Onsager reciprocity (16).

LIGHT-INDUCED THERMAL- AND BAROEFFECT

The Onsager coefficients (15) can be used to estimate pressure and temperature gradients occurring in a closed and insulated system. Closed insulated capillary is an example of such a system. Resonant electromagnetic radiation with fixed force X_S causes pressure and temperature drops at the ends of the capillary. Thermodynamic forces form a numerical flow J_p and heat flux J_T in a capillary. According to Prigogine theorem [11], in a closed and insulated system these fluxes are equal to zero:

$$\begin{aligned} J_p &= \Lambda_{pp} X_p + \Lambda_{pT} X_T + \Lambda_{ps} X_s = 0, \\ J_T &= \Lambda_{Tp} X_p + \Lambda_{TT} X_T + \Lambda_{Ts} X_s = 0. \end{aligned} \tag{18}$$

From Eqs. (18) we obtain:

$$\begin{aligned} X_p &= -k_Б \nu = X_s\left(\Lambda_{pT}\Lambda_{Ts} - \Lambda_{ps}\Lambda_{TT}\right)/\Delta_0, \\ X_T &= -\frac{\tau}{T_0} = X_s\left(\Lambda_{Tp}\Lambda_{ps} - \Lambda_{pp}\Lambda_{Ts}\right)/\Delta_0, \\ \Delta_0 &= \Lambda_{pp}\Lambda_{TT} - \Lambda_{Tp}\Lambda_{pT}. \end{aligned} \tag{19}$$

The coefficients Λ_{ik} can be calculated if the perturbation functions h_i are known.

Analytical expressions for the kinetic coefficients can be easily obtained for the free molecular regime. We neglect inelastic collisions of the gas particles with the wall. As boundary conditions let us assume specularly-diffuse model, according to which the some fraction $(1-\alpha_i)$ of the particles of *i*th species is reflected specularly, and fraction α_i of the particles is scattered diffusely with the Maxwell velocity distribution function.

The theory is linear in $\chi(\mathbf{v})$. Therefore we can assume that $n_m << n_n$, i.e. $n_n \approx n$. The experiments carried out by [12] show that the coefficients of specularly-diffuse reflection for excited and unexcited particles are slightly different. Thus, in the theory small parameter can be introduced:

$$|\Delta\alpha| = |\alpha_n - \alpha_m| << 1. \tag{20}$$

where α_m, α_n are the accommodation coefficients of excited and unexcited atoms respectively.

Inhomogeneous broadening of the absorption line ($\Gamma << k_B\overline{v}$) is typical for rarefied gases. In this case using small parameter (20) for the free molecular regime we obtain the expressions for the pressure and temperature gradients

$$\begin{aligned} \frac{dp}{dz} &= \frac{10\pi^{3/2}}{\lambda}\left(\frac{\Omega}{k\overline{v}}\right)\left(\frac{G_{mn}}{k\overline{v}}\right)^2 \exp\left[-\left(\frac{\Omega}{k\overline{v}}\right)^2\right] p\Delta\alpha \\ \frac{dT}{dz} &= -\frac{12\pi^{3/2}}{\lambda}\left(\frac{\Omega}{k\overline{v}}\right)\left(\frac{G_{mn}}{k\overline{v}}\right)^2 \exp\left[-\left(\frac{\Omega}{k\overline{v}}\right)^2\right] T\Delta\alpha. \end{aligned} \tag{21}$$

Here λ is the wavelength of *m*-*n* transition.

RESULTS

. The pressure and temperature gradients depend on the difference of accommodation coefficients of atoms in the excited and ground state $\Delta\alpha$, on the ratio of the detuning Ω to the Doppler shift $k\overline{v}$ and intensity of the radiation I. Gradients reach their maximum values at $\Omega/k\overline{v} = 0{,}7$. In the exact resonance at $\Omega = 0$ light–induced pressure and temperature gradients are not available, as the LID and LIHT [1, 5]. Gradients directions are determined by the signs of detuning from the center of the absorption line and the difference of the accommodation coefficients.

In the free molecular regime pressure and temperature gradients have in opposite directions at all conditions. Fig.1 shows a picture of the gradients generated in the closed insulated capillary. If $\Omega > 0$ and $\Delta\alpha < 0$, LID occurs and gas as a whole will move in the direction of movement of the unexcited *n*-component. The pressure gradient opposite to the propagation of radiation appears in the capillary. According to [5], a surface light-induced drift and the heat flux in the whole range of Knudsen numbers are directed opposite to each other. In a closed insulated system this leads to the establishment of oppositely pressure and temperature gradients in the free molecular regime.

Let us make numerical estimates for the gradients of sodium vapor (the wavelength of transition is $\lambda = 5893\,\text{Å}$). Let the intensity of the electromagnetic wave be equal to $I = 1\,W/cm^2$. If the transition dipole moment $d_{mn} = 2\,\text{D}$, the Rabi frequency is equal to $G_{mn} = 10^8\,Hz$; Doppler broadening is $k\overline{v} = 2\pi\overline{v}/\lambda = 5\cdot 10^9\,Hz$, $\Omega/k\overline{v} = 0{,}3$. Typical characteristic for the atoms are: $|\Delta\alpha| = 0{,}001$. We assume that the radius of the capillary is equal to $r_0 = 3\,mm$ and the average temperature in the system is $T_0 = 350\ K$. The pressure is $p = 0{,}045\,Pa$. In this case from (21) we obtain

$$\frac{dp}{dz} = 0{,}5\ \frac{Pa}{m}, \qquad \frac{dT}{dz} = -4{,}35\cdot 10^3\ \frac{K}{m}. \tag{22}$$

The saturation effect leads not only to a strong heating of the gas in the optical wavelength range [13], but also to a large temperature gradient in a closed insulated system. In the nearly free molecular regime where the collisions between particles are beginning to emerge, the temperature difference disappears, and the temperature gradient begins to decrease.

CONCLUSION

Cross-transfer processes in a one-component gas due to resonant laser radiation and the gradients of pressure and temperature were studied. Based on a system of kinetic equations in the linear approximation, the expression for the entropy production is obtained. It is shown that the kinetic coefficients determining transfer processes satisfy the Onsager reciprocity in all Knudsen numbers and arbitrary interaction of gas atoms with the surface of the capillary. Using Prigogine theorem of stationary weakly nonequilibrium states, light- induced baroeffect and thermaleffect were considered. For free molecular regime numerical estimates for pressure and temperature gradients emerging in a closed insulated capillary have been made. Found that the gradients at inhomogeneous broadening of the absorption line in the free molecular regime are determined by the difference of accommodation coefficients of excited and nonexcited atoms, the value of the detuning and intensity of the radiation. The temperature gradient in this regime takes very high values and is directed opposite to the pressure gradient.

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